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IMPROVING THE QUALITY OF PYROLYSIS OIL FROM CO-FIRING HIGH-DENSITY POLYETHYLENE PLASTIC WASTE AND PALM EMPTY FRUIT BUNCHES

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ABSTRACT

This study aimed to produce and improve the quality of pyrolysis oil as a source of bioenergy that is made by mixing palm empty fruit bunch (EFB) with high-density polyethylene (HDPE) plastic waste. The slow co-pyrolysis method was employed, and HDPE waste and EFB were fed into the pyrolysis reactor at HDPE amounts of 0, 10, 25, 50, 75, and 100% by weight. The pyrolysis oil product was obtained by co-firing EFB with HDPE using the slow co-pyrolysis method in a fixed bed reactor at 500°C with a flow rate of 750 mL/min and a heating rate of 5°C/min. The chemical compositions of pyrolysis oil were analyzed by gas chromatography-mass spectroscopy. A pyrolysis oil produced by HDPE 100 wt.% was dominated by the chemical compounds of phenols, aromatics, aliphatic, and acids, while for EFB 100 wt.% was dominated with aldehydes, acids, phenols, furan and aliphatic. The addition of HDPE reduced the amount of pyrolysis oil yield, increased the pH, reduced the viscosity, and reduced the oxygen content of the pyrolysis oil. These results proved that the HDPE affected the decrease in pyrolysis oil and the increase in gas production from co-firing HDPE and EFB using the slow co-pyrolysis method.

Keywords: Bioenergy; HDPE; Improving the quality; EFB; Pyrolysis oil; Reduced the oxygen

1. INTRODUCTION

Development of new technologies to generate energy from renewable biomass is currently considered a promising alternative fuel source, as fossil fuel supplies are diminishing and their use is increasing carbon dioxide emissions. Various catalytic chemical processes have been proposed to utilize biomass (Pidko et al., 2010; Zhang et al., 2011; Degirmenci et al., 2011a, 2011b; Degirmenci et al., 2014; Oozeerally et al., 2018; Kusrini et al., 2018). However, thermochemical conversion remains one of the most attractive methods because of its compatibility with existing industrial chemical processes. In this respect, pyrolysis is a method of short-chain decomposition through a high-temperature heating process without the use of oxygen, which generates a range of products, namely biochar, gases, and vapors (condensed to obtain pyrolysis oil) (Oudenhoven et al., 2016). The pyrolysis method has economic advantages

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because it does not require pre-treatment of the feedstock.

Pyrolysis oil is an important alternative energy resource, as it has the potential to replace fossil fuels with a renewable resource to diminish greenhouse gas emissions and at the same time to increase the value of agricultural and plastic waste. Therefore, pyrolysis oil could create an additional economic value for poor communities, such as rural populations and individuals who recycle plastic waste (Sukiran et al., 2017). Pyrolysis oil can be synthesized from forestry biomass, crop residues (agricultural biomass) such as empty fruit bunches (EFB), palm kernel shell, mesocarp fiber, oil palm frond, corn cobs and oil palm trunk (Das et al., 2009; Supramono et al., 2016a; Sukiran et al., 2017). An advantage of agricultural biomass is that it contains less sulfur and is abundantly available as a feedstock.

EFB is the main agricultural waste of the palm industry, and it is an abundant waste biomass that is harvested in tropical regions such as in Indonesia and Malaysia (Purwanto et al., 2015; Ro et al., 2018). EFB quantities have been increasing in recent years due to the increasing cultivation of palm. Although EFB can be returned to the soil as fertilizer or burnt to generate steam for electricity production, most EFB is dumped in landfill because of its low economic value. Thus, a large amount of EFB is currently not utilized and can be used as a feedstock for pyrolysis oil production (Chang, 2014; Purwanto et al., 2015). The oil is obtained from the pyrolysis of EFB at approximately 500°C in a fluidized bed reactor; the calorific value usually is approximately 20 MJ/kg (Sukiran et al., 2017). A similar observation has been reported for pyrolysis of biomass with heating at temperatures of 450–550°C (Oudenhoven et al., 2016). As has been recently reported, rapid pyrolysis that involves brief contact time is a promising method (Das et al., 2009; Kim et al., 2011). The co-pyrolysis method is a more promising way to convert the waste into higher-value oil (Gang & Aimin, 2008). It can reduce the phase separation of pyrolysis oil, and thus it can improve the stability of the oil (Supramono et al., 2016b). This technique can therefore reduce the volume of waste and achieve synergy by co-processing biomass with plastics and other materials (Gang & Aimin, 2008).

Through pyrolysis, environmental pollution could be reduced and the added value of agro-waste could be increased by energy generation mainly through the conversion of agricultural biomass together with plastic (high-density polyethylene [HDPE]) waste into energy. In addition, co-pyrolysis has an advantage over pyrolysis of either agricultural waste or plastic waste separately. The co-pyrolysis product yield is higher than when adding the individual pyrolysis oil produced separately by pyrolysis of agricultural or plastic waste, because of the interaction of free radicals during the thermal treatment process (Bhattacharya et al., 2009; Supramono et al., 2016b). The chemical composition of oil from waste agricultural biomass contains significant amounts of oxygenated organic compounds, and therefore results in a high O/C ratio, high moisture causing low energy density, and a low heating-value product. This also leads to high soot formation. Furthermore, the presence of oxygenated products reduces the stability and therefore the shelf life and storage duration of the oil (Sabil et al., 2013). The consumption of plastic has increased very quickly worldwide, making plastic waste very abundant (Bhattacharya et al., 2009). Increasing plastic demand leads to increasing waste accumulation every year (Sabil et al., 2013; Sharuddin et al., 2016). A significant percentage of plastic dumped in landfills are HDPE (57%) (Sogancioglu et al., 2017).

The direct use of pyrolysis oil from agricultural waste as a fuel is not viable because of the low heating value of pyrolysis oil. This is caused by the rich oxygen-containing compounds (Ro et al., 2018; Rodionova et al., 2017). Co-pyrolysis of EFB with plastic waste can reduce the oxygen compounds and increase the caloric value of pyrolysis oil. The addition of plastics in biomass pyrolysis increases the yield and the resulting oil calorific value compared with biomass pyrolysis alone, due to the presence of paraffin-containing hydrocarbon polymers,

isoparaffin, olefin, naphtha, and aromatics (Bhattacharya et al., 2009; Abnisa et al., 2014; Uzoejinwa et al., 2018).

In this study, we produced pyrolysis oil by co-firing EFB with HDPE using a slow co-pyrolysis method at 500°C with a flow rate of 750 mL/min and a heating rate of 5°C/min using a fixed bed reactor. The EFB and HDPE as feedstock were located inside a metal half-cylindrical boat in a pyrolysis reactor. The characteristics of the pyrolysis oil product were analyzed in detail.

2. EXPERIMENT

2.1. Materials

HDPE waste was obtained from Jakarta, Indonesia, while EFB as biomass feed was obtained from Jambi (Sumatra, Indonesia). The EFB and HDPE waste were prepared by drying in an oven at 105°C to reduce the moisture content to less than 10 wt.%. The raw materials were then chopped with a grinder, and sizes of 1–3 cm were obtained.

2.2. Slow Co-pyrolysis in a Fixed Bed Reactor

The design of the reactor for the slow co-pyrolysis method followed that reported by Supramono et al. (2016b). Slow co-pyrolysis was done in a stainless steel reactor of a cylindrical type with a length of 25 cm and a diameter of 6 cm. Feedstock composed of EFB and HDPE were placed in a boat made of half-cylinder sizing with a length of 11 cm and a diameter of 5 cm. Each experiment used 20 g feeds (EFB and HDPE). The mass ratio of feedstock HDPE waste to EFB was varied to 0:100, 25:75, 50:50, 75:25, and 100:0. The boat was located horizontally inside the cylindrical reactor. Nitrogen flow was adjusted to 3 L/h in an up-flow reactor configuration to fulfill the maximum residence time of 7–10 seconds to avoid byproduct and/or secondary reaction of the plastic. Prior to pyrolysis, the reactor was flushed with nitrogen gas for 15 min with a flow rate of 750 mL/min. The heating rate of the pyrolysis process was maintained and set at 5°C/min to heat the nitrogen gas at 500°C. After reaching 500°C, the temperature was maintained for 30 min for each run to produce more yield of pyrolysis oil product. After that, the vapors product was delivered to the condenser by cooling water at 10°C to allow the vapor product to become liquid fraction (pyrolysis oil). The pyrolysis oil was collected in an Erlenmeyer flask and kept for further characterizations. The reactor tube was then cooled to room temperature after each slow co-pyrolysis run, the solid fraction product was collected. The non-condensable gases with compositions of H_2, CO, CO_2 and a small of hydrocarbon was not collected.

2.3. Characterization

Volatile matter was measured in accordance with ASTM standard no. 3175, in which the size of the EFB is approximately 1–3 cm and the weight is 2 g. It was then put in a porcelain crucible and kept in a furnace with a temperature of 550°C for 10 min and weighed after cooling at room temperature, then kept in a desiccator.

The chemical composition of the pyrolysis oil product was analyzed using gas chromatography-mass spectrometry (NIST GCMS-QP1010 Ultra, Shimadzu, UK). The pyrolysis oil was extracted using diethyl ether solvent and concentrated with nitrogen gas. Thermal analysis of the EFB and HDPE were determined using a thermogravimetric (TG) analyzer (STA 6000, PerkinElmer). The viscosity of the pyrolysis oil was determined using a Saybolt universal viscometer at a temperature of 40°C. This viscosity was compared with that of commercial fuels.

3. RESULTS AND DISCUSSION

3.1. Materials

The water content, ash, solid carbon, heating value, and volatile matter in the feed were determined, and their relative contents are given in Table 1. The ultimate test was performed to determine the levels of C, N, O, and S. The results of proximate and ultimate analysis of the EFB and HDPE are listed in Table 1. The presence of water can reduce the heat value, especially the low heating value and combustion temperature.

However, water can increase the ignition delay or burning delay, and can also lower the combustion rate when compared with commercial fuel (Egorov et al., 2018). Water can also reduce the viscosity of pyrolysis oil product; therefore, it is more feasible for use as fuel. The oxygen content of the EFB is high enough to reach 24.84%, which causes high levels of oxygenated organic compounds in pyrolysis oil and low calorific value. The carbon content in the HDPE is 84.74%, while in EFB it is only approximately 45.94%. The higher volatile matter in the EFB (65.9 wt.%) and HDPE (89.9 wt.%) can result in higher pyrolysis oil yields. The vapour fraction will be condensed to be pyrolysis oil. High levels of ash content can cause pyrolysis oil to become corrosive and can reduce its durability. On the other hand, the ash content also influences the capability of external heating in the pyrolysis reactor (Supramono et al., 2016b). Since ash can absorb the heat, the pyrolysis process is not effective.

Table 1 Proximate and ultimate analysis of EFB and HDPE

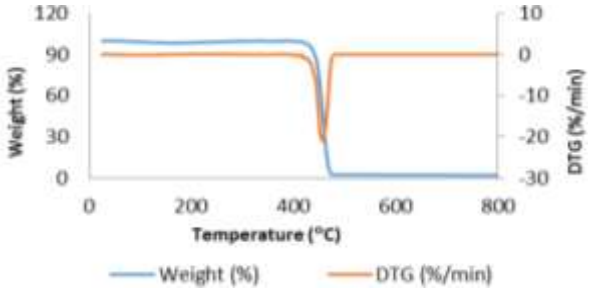
Materials	EFB (wt. %)	HDPE (wt. %)
Water	13.6	10.0
Ash	4.9	0.1
Volatile matter	65.9	89.9
Fixed carbon	15.6	0.02
Carbon	45.9	84.7
Hydrogen	5.6	11.6
Nitrogen	4.0	0.02
Sulfur	0.14	0.7
Oxygen	24.8	2.9

3.2. Thermogravimetric analysis

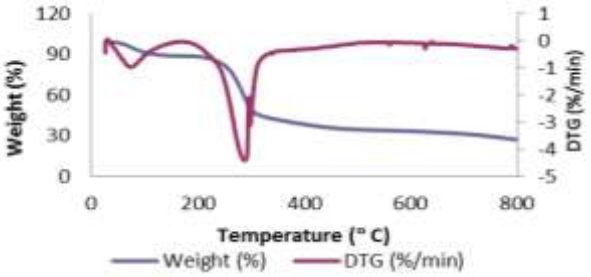
A thermogravimetric analyzer was used to monitor the mass loss profile of materials. The TG curve is the actual sample mass normalized with the initial dehumidified sample mass as a function of the temperature, while the derivative thermogravimetric (DTG) curve is the rate of mass change as a function of time or temperature (Gang & Aimin, 2008). DTG is commonly used to characterize the degradations and to establish kinetic parameters (Martí-Rosselló et al., 2018). Figures 1a–1c present the TG and DTG curves for the HDPE, EFB, HDPE, and their mixture as a function of temperature. The thermal behavior of the HDPE and EFB were studied in a range of 25°C to 800°C. The main thermal decomposition of the HDPE occurred at 454°C. On the other hand, the EFB began to decompose at 262°C and the main weight loss due to decomposition occurred at 436°C. The devolatilization process can be seen in the DTG curves. The HDPE showed only one peak at 454°C, while the EFB showed three peaks at 80°C, 283°C, and 294°C. In DTG for the EFB, the shoulder peak was located at a higher temperature. It can be identified as lignocellulosic biomass because EFB mainly consists of cellulose, hemicellulose, and lignin (Gang & Aimin, 2008; Martí-Rosselló et al., 2018). Based on these behaviors of the materials, the co-pyrolysis was conducted at a temperature of 500°C, which is higher than the decomposition temperature of both ingredients. We used a higher temperature for pyrolysis than those found in the slow co-pyrolysis (450°C) process for wood/HDPE reported by Bhattacharya et al. (2009).

3.3. Slow Co-pyrolysis of EFB and HDPE

There are three types of products of slow co-pyrolysis of the mixture HDPE and EFB: solid, vapor (condensed to be liquid), and gas. The amount of solid product increased when the EFB decreased from 100 to 90 wt.%. Similarly, when the EFB decreased from 75 to 0 wt.%, the amount of solid product also decreased (see Table 2). The amount of pyrolysis oil decreased from 17.4 to 6 wt.% and the amount of gas product increased from 43.6 to 72.5 wt.% when the mass ratio of the HDPE increased. These results proved that the HDPE affected the decrease in pyrolysis oil product and the increase in gas product.



(a)



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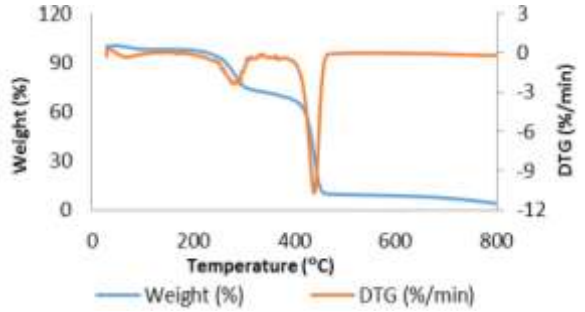

(c)

Figure 1 TG and DTG curves of (a) HDPE; (b) EFB, and (c) EFB and HDPE waste with mass ratio of 50:50

Characteristics and chemical composition of pyrolysis oil product with varying HDPE mass ratios in the feedstock are given in Table 3. The highest yield of pyrolysis oil was obtained for the HDPE ratio of 0 wt.%. As shown in Figure 2, the pyrolysis oil yield of the HDPE was about 17.4 %. The weight of gas products was greater than solid and liquid products.

The color of pyrolysis oil product from the EFB 100 wt.% was clear yellow (see Figure 3a), while that of pyrolysis oil produced by the HDPE was murky yellow (see Figure 3b). For pyrolysis oil product produced from a mixture of EFB and HDPE wastes, the resulting color was deep yellow (see Figure 3c). This is caused by different types of raw material and its components. This indicated that there are dyes in HDPE plastic waste that affect the color of the pyrolysis oil product, with the bio-oil from the EFB a reddish-brown color produced by the

presence of the catalyst Ni/ZSM-5 in the reactor using a rapid pyrolysis process (Purwanto et al., 2015). These colors are significantly different, probably due to the different method used for production of the pyrolysis oil.

Table 2 The three types of products from slow co-pyrolysis

Feedstock (wt.%)		Yield (wt.%)		
HDPE	EFB	Solid	Pyrolysis oil	Gas
0	100	39.00	17.40	43.60
10	90	47.00	16.35	36.65
25	75	36.20	15.90	47.90
50	50	40.65	13.00	46.35
75	25	37.40	6.75	55.85
100	0	21.50	6.00	72.50

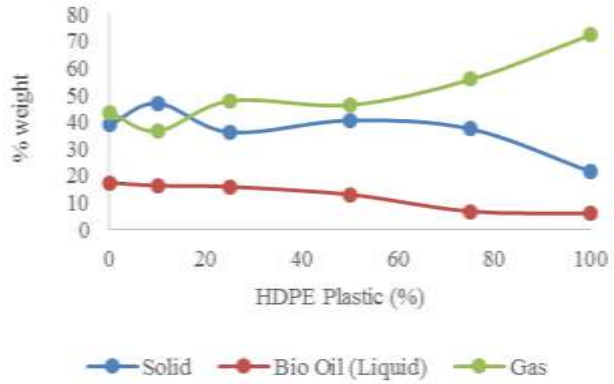

Figure 2 Distribution of solid, pyrolysis oil and gas fractions using slow co-pyrolysis of EFB and HDPE at 500°C

Table 3 Characteristics of pyrolysis oil from EFB and HDPE of varying feedstock ratios with feedstocks of 20 g

HDPE:EFB ratio (wt.%)	Yield of pyrolysis oil (g)	Yield pyrolysis oil (%)	Appearance	pH
0:100	3.48	17.40	clear	3
10:90	3.27	16.35	clear	3
25:75	3.18	15.90	cloudy	3
50:50	2.60	13.00	cloudy	3
75:25	1.35	6.75	cloudy	4
100:0	1.20	6.00	cloudy	5

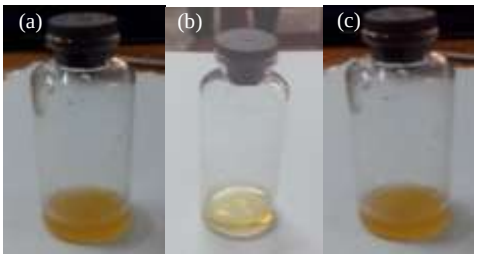

Figure 3 Color of pyrolysis oil products for: (a) HDPE 100 wt.%; (b) EFB 100 wt.%; and (c) EFB:HDPE is 50:50 wt.%

3.4. Pyrolysis Oil Product Distribution and Characterization

The chemical compositions of the pyrolysis oil from slow co-pyrolysis method were analyzed by GC-MS, and more than 50 compounds were identified. The compounds identified in the

pyrolysis oil include aromatics, aliphatic, phenols, furans, alcohols, aldehydes, ketones, and acids. In addition to these compounds, there are also negligible complex compounds of hydrogen. According to the literature review, EFB can be composed into cellulose, hemicellulose, and lignin (Gang & Aimin, 2008). According to the pyrolysis process, the cellulose can produce levoglucosan, which would then be changed into aromatic and phenol compounds in the presence of a catalyst, while hemicellulose can be converted into phenol, ketone, and acid compounds, and lignin can form a phenolic complex (Stefanidis et al., 2014). Thus, it can be used as a reference to determine the composition of pyrolysis oil. The chemical compositions of pyrolysis oil products are shown in Tables 4–6. It was clearly observed that the presence of the HDPE waste in the pyrolysis process increased the heating value of pyrolysis oil, indicating an increase in carbon and hydrogen contents. A similar trend has also been reported for pyrolysis oil produced from wood/plastics materials in a lab-scale auger-fed reactor at 1 atm with a short vapor residence time (Bhattacharya et al., 2009). This suggests that the slow co-pyrolysis method was successfully designed to produce pyrolysis oil through HDPE plastic thermal decomposition. The energy is enough to produce vapor rapidly and to prevent the vaporized HDPE from condensing on EFB chars.

GC-MS characterization showed that the chemical composition of pyrolysis oil produced by HDPE 100.wt% was dominated by the major components of phenols, aromatics, aliphatic, and acids (see Figure 4). While for EFB 100 wt.%, the chemical composition of pyrolysis oil is dominated with aldehydes, acids, phenols, furan and aliphatic (see Figure 4).

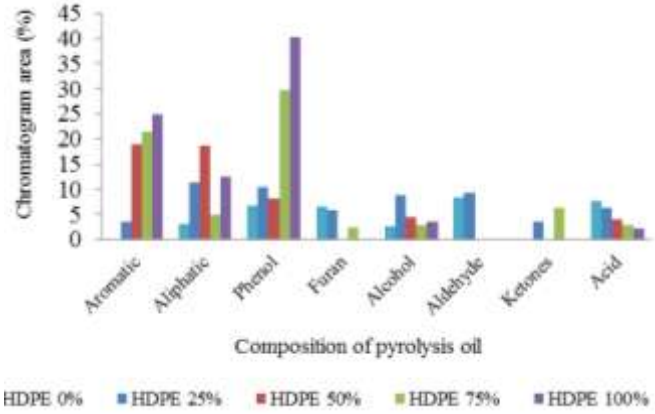

Figure 4 The chemical composition of pyrolysis oil product with different mass ratios of HDPE plastic waste

Table 4 Major chemical compositions of pyrolysis oil with EFB:HDPE mass ratio of 100:0

Ret. Time	Area	Area (%)	Name
8.97	434202	10.06	Phenol, 2-methoxy-
6.23	339268	7.88	Phenol
16.09	246020	5.71	Phenol, 2,6-dimethoxy-
7.36	181151	4.21	1,2-Cyclopentanedione, 3-methyl-
10.89	180199	4.18	Phenol, 4-ethyl-
5.23	171037	3.97	1,2-Cyclopentanedione
14.14	141851	3.29	Phenol, 4-ethyl-2-methoxy-
12.31	139684	3.24	Benzofuran, 2,3-dihydro-
8.44	135246	3.14	Phenol, 3-methyl-
5.99	131198	3.05	2-Furancarboxaldehyde, 5-methyl-
8.19	129218	3.00	Oxirane, propyl-
7.98	115535	2.68	N-(2-Furylcarbonyl)tyrosine
26.60	113447	2.63	Phenol, 2,6-dimethyl-4-nitro-
17.45	107372	2.49	Vanillin
15.11	103039	2.39	2-Methoxy-4-vinylphenol

Table 5 Major chemical composition of pyrolysis oil product with EFB:HDPE mass ratio of 0:100

Ret. Time	Area	Area (%)	Name
15.10	64682485	12.58	1-.beta.-d-Ribofuranosyl-1,2,4-triazole-3-carboxylic acid
15.58	54028274	10.51	Phenol, 4-ethyl-2-methoxy-
15.32	29682357	5.77	Benzofuran, 2,3-dihydro-
26.67	26321013	5.12	1-Hexacosene
26.03	24729421	4.81	3-Chloropropionic acid, tridecyl ester
8.86	22832404	4.44	2-Furancarboxaldehyde, 5-methyl-
20.85	20013550	3.89	1,22-Docosanediol
22.66	18191079	3.54	Heneicosane
8.97	18086141	3.52	2-Cyclopenten-1-one, 3-methyl-
27.68	14534588	2.83	11-Hexadecen-1-ol, acetate, (Z)-
21.52	13304534	2.59	17-Pentatriacontene
27.39	10858942	2.11	1-Dodecanol, 2-hexyl-
17.00	10535949	2.05	Vanillin, acetate
21.07	9375926	1.82	1,3-Dimethyl-(3,7-dimethyloctyl)cyclohexane
27.15	9335264	1.82	Cyclodocosane, ethyl-

Table 6 Major chemical composition of pyrolysis oil product with EFB:HDPE mass ratio of 50:50

Ret. Time	Area	Area (%)	Name
6.564	983210	26.4	Phenol
7.129	129304	3.47	2-Furanmethanol, tetrahydro
9.246	338436	9.08	Phenol, 2-methoxy-
16.448	175662	4.71	Phenol, 2,6-dimethoxy-
28.895	169853	4.56	3-Pyridinecarboxylic acid, 1,2-dihydro-4-hydroxy-6-methyl-2-oxo-, ethyl ester
28.950	184179	4.94	1,2-Octadecanediol
28.999	359191	9.64	Guanidine, N-[3-[(2-bromophenyl)amino]-1-propenyl]-
29.156	527171	14.15	2- Bromopropionic acid, pentadecyl ester
29.180	266924	7.16	1-Hydroxy-4-methoxyiminomethyl-2,2,5,5-tetramethyl-3-imidazoline-3-oxide
29.851	404112	10.85	2- Bromopropionic acid, pentadecyl ester
29.880	187831	5.04	.alpha.-D-Glucopyranoside, methyl 4,6-O nonylidene-

3.5. Viscosity and acidity

The viscosity of the pyrolysis oil product without HDPE plastic waste was 7 cSt, and it decreased to 3.6 and 4.8 cSt with 50 and 100 wt.% HDPE plastic waste, respectively. All pyrolysis oil has a higher viscosity than commercial fuels. The presence of high lignin may cause the pyrolysis oil to become more viscous, making the product unstable. The viscosity influences the flow ability of pyrolysis oil and ignition delay time (Supramono et al., 2016b), when used as diesel oil.

The pH of pyrolysis oil produced from 100 wt.% EFB was 3. This is because pyrolysis oil contains many oxygenated compounds, which not only change the color of the pyrolysis oil but also produce radicals. With its high acidity, the pyrolysis oil from 100 wt.% EFB cannot be used as a fuel. It has been reported in the literature that the bio-oil product from woody biomass is unstable during storage and corrosive because of its organic acid content and has a low heat content and high density compared with petroleum fuel (Bhattacharya et al., 2009). On the other hand, the pH of pyrolysis oil produced from 100 wt.% HDPE is 5 due to the absence of oxygenated compounds, making it resilient to oxidization. Overall, this finding implies that

pyrolysis oils produced by various mixture of HDPE and EFB have different pH values between the two extremes of pH 3–5.

4. CONCLUSION

In this work, we have investigated the impact of co-pyrolysis of plastic waste (HDPE) with agricultural waste (EFB) for pyrolysis oil production. We demonstrated that the addition of HDPE waste in EFB reduces the pyrolysis oil yield. The pH of pyrolysis oil is increased with lower EFB, which is an advantage for its further use.

5. ACKNOWLEDGEMENT

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